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**METHODS AND SYSTEMS FOR  
ACOUSTICALLY-ASSISTED  
HYDROPROCESSING AT LOW PRESSURE**

STATEMENT REGARDING FEDERALLY  
SPONSORED RESEARCH OR DEVELOPMENT

This invention was made with United States Government support under contracts DE-AC0576RLO1830 and HR0011-10-C-0088 to Battelle Memorial Institute. The Government has certain rights in this invention.

**BACKGROUND**

Hydroprocessing is important to the production of a wide range of chemicals and fuels. Hydroprocessing units can have many forms and purposes within a refinery and are often the most common processing and upgrading units. For example, hydrotreating capacity within a refinery can comprise over 55% of the total refining capacity. Hydroprocessing can also be one of the most energy intensive and costly processes of fuel and chemical processing facilities. One of the reasons for the high energy usage and cost, is the high hydrogen pressure, often greater than 68 psi, required by traditional techniques.

Generally hydroprocessing is the catalytic conversion and removal of organic heteroatoms (sulfur, nitrogen, oxygen, and metals) and partial, or full, saturation of aromatic hydrocarbons from various refinery streams. As the quality of crude oils decrease and the production of fuels and chemicals from alternative feedstocks (i.e., biomass, coal, oil shales, tar sands, etc.) increases, the hydroprocessing demand within the refinery will have to increase to meet the increasingly stringent environmental regulations placed on fuel specifications and the growing world-wide demand for diesel fuels. Furthermore as more alternative oils and heavy and extra-heavy oils enter the market, not only would hydrotreating demands increase for removal of heteroatoms and aromatics, but hydrocracking capacity can be expected to grow to increase production of gasoline, jet, and diesel fuels from heavier hydrocarbon fractions, such as vacuum gas oils (VGO), petroleum resid (bp>550° C.), and bitumen. Accordingly, a need exists for low pressure hydroprocessing methods and systems.

**SUMMARY**

This document describes methods and systems for hydroprocessing at relatively low pressures. In one embodiment, a method for hydroprocessing a feedstock comprising one or more hydrocarbon compounds carried in, or mixed with, a transport gas is characterized by flowing the feedstock through a reaction zone in a reactor that has a bulk pressure less than 68 atm and applying acoustic energy into and/or through the reaction zone. The method further comprises chemically reacting the hydrocarbon compounds with a hydrogen source in the presence of a catalyst, wherein the reacting occurs in the reaction zone.

As used herein, hydroprocessing can refer to the general processes of hydrotreating, hydrogenation, and/or hydrocracking of hydrocarbon sources for the production of fuels and chemicals. Hydrotreating can refer to a collection of processes that include hydrodesulfurization (HDS), hydrodenitrogenation (HDN), hydrodeoxygenation (HDO), hydrodemetalization (HDM), or hydrodehalogenation for the removal of heteroatoms from hydrocarbon mixtures containing organic heteroatoms. Hydrotreating is a relatively mild hydroprocessing process that aims to selectively hydrogenate

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and remove the heteroatomic atom(s) from the organic heteroatoms, without significantly altering the boiling range of the final product.

Hydrogenation processes can refer to processes in which the raw hydrocarbons, typically containing aromatics, olefins, or organic heteroatoms are partially or fully saturated with hydrogen to alter the chemical composition and/or properties of the feedstock without complete removal of the heteroatom or ring-opening or molecular size reduction of aromatics or polyaromatics. Examples of hydrogenation processes can include, but are not limited to aromatic saturation to naphthenes; saturation of alkenes or alkynes to their respective paraffin (or naphthene); saturation of aldehydes, ketones, epoxides, or organic acids to alcohols; saturation of nitriles, anilines, or imines to amines; saturation of nitro groups to anilines, amines, or oximes; or related hydrogen saturation processes.

Hydrocracking can refer to processes in which hydrogenation and/or hydrotreating occur and are accompanied by cracking or cleavage of carbon-carbon bonds within the parent molecule. Examples include hydrocracking of long-chain paraffins to smaller paraffins; ring-opening of naphthenes, saturation of polyaromatics to naphthenes followed by ring-opening to generate alkyl naphthene and/or paraffins; dealkylation of aromatics and/or naphthenes; demethylation (or alkylation) of iso-paraffins to n-paraffins, and other related hydrotreating or hydrogenation processes where the molecular size/weight of the parent molecule is reduced.

In some embodiments, the acoustic energy induces non-linear acoustic effects (e.g. acoustic streaming and radiation pressure) that, for example, can enhance mass transfer (e.g. mixing, infiltration, extraction, etc.). Non-linear acoustic energy can be generated using devices that provide acoustic frequencies and sound pressure levels that are together sufficient to induce these effects. In but one example, at 20 kHz, sound pressure levels greater than 100 dB can produce non-linear acoustic effects in predominantly gas-based hydroprocessing streams. Calculations of the acoustic Reynolds numbers for 20 kHz at sound pressure levels of 150-170 dB in the gases tested yielded values of ~1 to 200. Reynolds numbers greater than 1 can induce acoustic streaming to enhance mass transfer in gas phase systems. Acoustic streaming is one example of a non-linear acoustic effect and is believed to be a primary acoustic mechanism responsible for enhancing mass and heat transfer. Acoustic streaming, as used herein, can refer to a steady flow field superimposed upon the oscillatory motion of a sound wave propagating in a fluid. It can occur due to the presence of boundaries or because of damping of the wave (attenuation). These steady flows have rotational character and their velocity increases with sound intensity.

The hydrocarbon compound can comprise solid particulates, liquid fluid and/or vapor. In preferred embodiments, the hydrocarbon compound is often, though not always, a liquid at room temperature and pressure; it can be in either the vapor or liquid phase under the reacting conditions. Examples of hydrocarbon compounds can include, but are not limited to, derivatives or distillate cuts of oils, tars, and/or asphaltenes. Examples of oil, tars, and/or asphaltenes can include, but are not limited to, petroleum, coal-derived oils, biomass-derived oils, oil sands, oil shale, and bitumen sources.

In some embodiments, the reaction zone can have a bulk pressure less than 34 atm. In others, the bulk pressure can be less than 17. In yet others, the bulk pressure can be less than 7 atm. In still others, the bulk pressure can be less than 3 atm. In preferred embodiments, the bulk pressure is approximately equivalent to atmospheric pressure. The liquid hourly space velocity (LHSV) can be greater than 0.1 hr<sup>-1</sup>.